60

What is claimed is:

1. An elastomeric composition usable for the manufacture of tires, based on a diene elastomer, an inorganic filler and a coupling agent comprising a polysilylated organosilicon compound which is at least bifunctional and can be grafted on to the elastomer by means of a sulfur group having a polythiosulfenamide function, of formula:

(I)
$$\equiv Si - A - S_x - N R^1 R^2$$

in which:

- A is a divalent bond group, whether straight-chain or branched, which makes it possible to join the polythiosulfenamide group to a first silicon atom of the organosilicon compound;
- x is an integer or fractional number of from 2 to 4;
- R^1 represents hydrogen, a monovalent hydrocarbon group, or R^2 ; and R^2 represents the grouping:

$$-B - Si \equiv$$
,

- B is a divalent bond group, whether straight-chain or branched;
- Si represents a second silicon atom of the organosilicon compound.
- 2. The composition according to Claim 1, wherein R^1 is selected from the group consisting of hydrogen, C_1 - C_8 alkyls, C_5 - C_{10} cycloalkyls, C_6 - C_{18} aryls, $(C_6$ - $C_{18})$ aryl- $(C_1$ - $C_8)$ alkyls, and R^2 .

- 3. The composition according to Claim 2, wherein R¹ is selected from the group consisting of hydrogen, methyl, ethyl, propyl, isopropyl, butyl, hexyl, benzyl, cyclohexyl, phenyl, and R².
- 4. The composition according to Claim 1, wherein R² represents the grouping:

$$-B - Si R^{3}_{(3-a)} (OR^{4})_{a}$$
,

- R³ represents a monovalent hydrocarbon group;
- R^4 represents hydrogen or a monovalent hydrocarbon group, which may be identical to or different from R^3 ; and
 - a is an integer equal to 1, 2 or 3.
- 5. The composition according to Claim 4, wherein the radicals R^3 and R^4 are selected from the group consisting of C_1 - C_8 alkyls, C_5 - C_{10} cycloalkyls, and phenyl.
- 6. The composition according to Claim 5, wherein the radicals R^3 and R^4 are selected from among C_1 - C_4 alkyls.
- 7. The composition according to Claim 1, wherein A and B, which may be identical or different, represent a hydrocarbon group comprising from 1 to 18 carbon atoms and optionally, one or more heteroatoms.
- 8. The composition according to Claim 7, wherein A and B, which may be identical or different, are selected from the group consisting of C_1 - C_{18} alkylenes and C_6 - C_{12} arylenes.
- 9. The composition according to Claim 8, wherein the sulfur group satisfies the formula:

(II)
$$\equiv Si - Z - S_x - NR^1 - Z - Si \equiv ,$$

wherein the groupings Z, which may be identical or different, represent a C₁-C₈ alkylene.

10. The composition according to Claim 8, wherein the sulfur group satisfies the formula:

(III)
$$\equiv Si - Z - S_x - NR^1 - S_y - Z - Si \equiv ,$$

wherein the groupings Z, which may be identical or different, represent a C_1 - C_8 alkylene, and y, which may be identical to or different from x, is an integer or fractional number from 2 to 4.

- 11. The composition according to Claim 1, wherein the diene elastomer is selected from the group consisting of polybutadienes, synthetic polyisoprenes, natural rubber, butadiene copolymers, isoprene copolymers and mixtures of these elastomers.
- 12. The composition according to Claim 1, wherein said composition comprises between 10 and 200 phr (parts by weight per hundred parts of elastomer) of reinforcing inorganic filler.
- 13. The composition according to Claim 1, wherein the quantity of coupling agent is between 1 and 20 phr.
- 14. The composition according to Claim 1, wherein the organosilicon compound is a silane-polythiosulfenamide of formula:

(IV)
$$(R^6O)_b R^5_{(3-b)} Si - A - S_x - N R^1 - B - Si R^3_{(3-a)} (OR^4)_a$$
,

in which:

- R⁵ represents a monovalent hydrocarbon group;
- R⁶ represents hydrogen or a monovalent hydrocarbon group, which may be identical to or different from R⁵;

b is an integer equal to 1, 2 or 3; and

- R⁵, R⁶ and b possibly being, respectively, identical to or different from R³, R⁴ and a.

15. The composition according to Claim 14, wherein the silane satisfies the formula:

(V)
$$(R^6O)_b R^5_{(3-b)} Si - Z - S_x - N R^1 - Z - Si R^3_{(3-a)} (OR^4)_a$$
,

in which the groupings Z, which may be identical or different, represent a C₁-C₄ alkylene, the radicals R³, R⁴, R⁵ and R⁶ representing a C₁-C₃ alkyl.

- 16. The composition according to Claim 15, wherein Z is propylene, R³, R⁴, R⁵ and R⁶ are selected from among methyl and ethyl.
- 17. The composition according to Claim 14, wherein the silane satisfies the symmetrical formula:

(VII)
$$[(R^4O)_a R^3_{(3-a)} Si - Z - S_x -]_2 N R^1$$

- 18. The composition according to Claim 17, wherein Z is propylene, R³, R⁴, R⁵ and R⁶ are selected from among methyl and ethyl.
- 19. The composition according to Claim 15, wherein x is integer or fractional number of from 2 to 3.
- 20. The composition according to Claim 19, wherein x is equal to 2.
- 21. The composition according to Claim 14, wherein the silane satisfies the formula:

(VI)
$$(R^6O)_b R^5_{(3-b)} Si - Z - S_x - N R^1 - S_y - Z - Si R^3_{(3-a)} (OR^4)_a$$

in which the groupings Z, which may be identical or different, represent a C_1 - C_4 alkylene, the radicals R^3 , R^4 , R^5 and R^6 represent a C_1 - C_3 alkyl, and y, which may be identical to or different from x, is an integer or fractional number of from 2 to 4.

22. The composition according to Claim 21, wherein Z is propylene, R^3 , R^4 , R^5 and R^6 are selected from among methyl and ethyl.

- 23. The composition according to Claim 21, wherein x and y are integers or fractional numbers of from 2 to 3.
- 24. The composition according to Claim 23, wherein x and y are equal to 2.
- 25. The composition according to Claim 14, wherein R¹ is selected from the group consisting of hydrogen, methyl, ethyl, propyl, isopropyl, hexyl, benzyl, cyclohexyl and phenyl.
- 26. The composition according to Claim 1, wherein the reinforcing inorganic filler is silica.
- 27. A process for preparing an elastomeric composition usable for the manufacture of tires, comprising the following steps:
- incorporating in a diene elastomer, in a mixer, at least:
 - a reinforcing inorganic filler;
- a coupling agent comprising a polysilylated organosilicon compound which is at least bifunctional and can be grafted onto the elastomer by means of a sulfur group, by
- thermomechanically kneading the entire mixture, in one or more stages, until a maximum temperature of between 110°C and 190°C is reached;
- cooling the entire mixture to a temperature of less than 100°C;
- then incorporating a vulcanization system; and
- kneading the entire mixture until a maximum temperature of less than 120°C is reached,
 wherein said sulfur group comprises a polythiosulfenamide function and satisfies the formula:

(I)
$$\equiv Si - A - S_x - N R^1 R^2,$$

-A is a divalent bond group, whether straight-chain or branched, which makes it possible to join the polythiosulfenamide group to a first silicon atom of the organosilicon compound;

- x is an integer or fractional number of from 2 to 4;
- R¹ represents hydrogen, a monovalent hydrocarbon group or R²; and

R² represents the grouping:

$$-B - Si \equiv$$
,

in which:

- B is a divalent bond group, whether straight-chain or branched;
- Si represents a second silicon atom of the organosilicon compound.
- 28. The process according to Claim 27, wherein R² represents the grouping:

$$-B - Si R^{3}_{(3-a)} (OR^{4})_{a}$$
,

in which:

- R³ represents a monovalent hydrocarbon group;
- R⁴ represents hydrogen or a monovalent hydrocarbon group, which may be identical to or different from R³; and
- a is an integer equal to 1, 2 or 3.
- 29. The process according to Claim 28, wherein the organosilicon compound is a silane polythiosulfenamide of formula:

(IV)
$$(R^6O)_b R^5_{(3-b)} Si - A - S_x - N R^1 - B - Si R^3_{(3-a)} (OR^4)_a$$
,

- R⁵ represents a monovalent hydrocarbon group;
- R⁶ represents hydrogen or a monovalent hydrocarbon group, which may be identical to or different from R⁵;
- b is an integer equal to 1, 2 or 3; and
- R⁵, R⁶ and b being, respectively, identical to or different from R³, R⁴ and a.
- 30. The process according to Claim 29, wherein the silane satisfies the formula:

(V)
$$(R^6O)_b R^5_{(3-b)} Si - Z - S_x - N R^1 - Z - Si R^3_{(3-a)} (OR^4)_a$$
,

in which the groupings Z, which may be identical or different, represent a C_1 - C_4 alkylene, and the radicals R^3 , R^4 , R^5 and R^6 represent a C_1 - C_3 alkyl.

31. The process according to Claim 29, wherein the silane satisfies the formula:

(VI)
$$(R^6O)_b R^5_{(3-b)} Si - Z - S_x - N R^1 - S_y - Z - Si R^3_{(3-a)} (OR^4)_a$$

in which the groupings Z, which may be identical or different, represent a C_1 - C_4 alkylene, the radicals R^3 , R^4 , R^5 and R^6 represent a C_1 - C_3 alkyl, and y, which may be identical to or different from x, is an integer or fractional number from 2 to 4.

32. A tire comprising an elastomeric composition based on a diene elastomer, an inorganic filler and a coupling agent comprising a polysilylated organosilicon compound which is at least bifunctional and can be grafted on to the elastomer by means of a sulfur group having a polythiosulfenamide function, of formula:

(I)
$$\equiv Si - A - S_x - N R^1 R^2,$$

-A is a divalent bond group, whether straight-chain or branched, which makes it possible to join the polythiosulfenamide group to a first silicon atom of the organosilicon compound;

- x is an integer or fractional number of from 2 to 4;
- R¹ represents hydrogen, a monovalent hydrocarbon group or R²; and

R² represents the grouping:

$$-B - Si \equiv$$

in which:

- B is a divalent bond group, whether straight-chain or branched;
- Si represents a second silicon atom of the organosilicon compound.
- 33. The tire according to Claim 32, wherein R² represents the grouping:

— B — Si
$$R^{3}_{(3-a)}(OR^{4})_{a}$$
,

in which:

- R³ represents a monovalent hydrocarbon group;
- R⁴ represents hydrogen or a monovalent hydrocarbon group, which may be identical to or different from R³; and
- a is an integer equal to 1, 2 or 3.
- 34. The tire according to Claim 33, wherein the organosilicon compound is a silane polythiosulfenamide of formula:

(IV)
$$(R^6O)_b R^5_{(3-b)} Si - A - S_x - N R^1 - B - Si R^3_{(3-a)} (OR^4)_a$$
,

- R⁵ represents a monovalent hydrocarbon group;
- R⁶ represents hydrogen or a monovalent hydrocarbon group, which may be identical to or different from R⁵:
- b is an integer equal to 1, 2 or 3;
- R⁵, R⁶ and b being, respectively, identical to or different from R³, R⁴ and a.
- 35. The tire according to Claim 34, wherein the silane satisfies the formula:

(V)
$$(R^6O)_b R^5_{(3-b)} Si - Z - S_x - N R^1 - Z - Si R^3_{(3-a)} (OR^4)_a$$
,

in which the groupings Z, which may be identical or different, represent a C_1 - C_4 alkylene, and the radicals R^3 , R^4 , R^5 and R^6 represent a C_1 - C_3 alkyl.

36. The tire according to Claim 34, wherein the silane satisfies the specific formula:

(VI)
$$(R^6O)_b R^5_{(3-b)} Si - Z - S_x - N R^1 - S_y - Z - Si R^3_{(3-a)} (OR^4)_a$$

in which the groupings Z, which may be identical or different, represent a C_1 - C_4 alkylene, the radicals R^3 , R^4 , R^5 and R^6 represent a C_1 - C_3 alkyl, and y, which may be identical to or different from x, is an integer or fractional number from 2 to 4.

37. A tire tread comprising an elastomeric composition based on a diene elastomer, an inorganic filler and a coupling agent comprising a polysilylated organosilicon compound which is at least bifunctional and can be grafted on to the elastomer by means of a sulfur group having a polythiosulfenamide function, of formula:

(I)
$$\equiv Si - A - S_x - N R^1 R^2,$$

-A is a divalent bond group, whether straight-chain or branched, which makes it possible to join the polythiosulfenamide group to a first silicon atom of the organosilicon compound;

- x is an integer or fractional number of from 2 to 4;
- R¹ represents hydrogen, a monovalent hydrocarbon group or R²; and

R² represents the grouping:

$$-B - Si \equiv$$

in which:

- B is a divalent bond group, whether straight-chain or branched;
- Si represents a second silicon atom of the organosilicon compound.
- 38. The tread according to Claim 37, wherein R² represents the grouping:

—.B — Si
$$R^{3}_{(3-a)}(OR^{4})_{a}$$
,

in which:

- R³ represents a monovalent hydrocarbon group;
- R^4 represents hydrogen or a monovalent hydrocarbon group, which may be identical to or different from R^3 ; and
- a is an integer equal to 1, 2 or 3.
- 39. The tread according to Claim 38, wherein the organosilicon compound is a silane polythiosulfenamide of formula:

(IV)
$$(R^6O)_b R^5_{(3-b)} Si - A - S_x - N R^1 - B - Si R^3_{(3-a)} (OR^4)_a$$
,

- R⁵ represents a monovalent hydrocarbon group;
- R⁶ represents hydrogen or a monovalent hydrocarbon group, which may be identical to or different from R⁵;

b is an integer equal to 1, 2 or 3; and

- R⁵, R⁶ and b being, respectively, identical to or different from R³, R⁴ and a.
- 40. The tread according to Claim 39, wherein the silane satisfies the formula:

(V)
$$(R^6O)_b R^5_{(3-b)} Si - Z - S_x - N R^1 - Z - Si R^3_{(3-a)} (OR^4)_a$$

in which the groupings Z, which may be identical or different, represent a C_1 - C_4 alkylene, and the radicals R^3 , R^4 , R^5 and R^6 represent a C_1 - C_3 alkyl.

41. The tread according to Claim 39, wherein the silane satisfies the specific formula:

(VI)
$$(R^6O)_b R^5_{(3-b)} Si - Z - S_x - N R^1 - S_y - Z - Si R^3_{(3-a)} (OR^4)_a$$

in which the groupings Z, which may be identical or different, represent a C_1 - C_4 alkylene, the radicals R^3 , R^4 , R^5 and R^6 represent a C_1 - C_3 alkyl, and y, which may be identical to or different from x, is an integer or fractional number from 2 to 4.